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## METHOD OF PRODUCING A COATING FOR ABSORBING NEUTRONS

The invention concerns a method of producing a coating to absorb neutrons created in the nuclear reaction of radioactive materials. The invention also concerns a shielding element produced by the method.

For the treatment of radioactive materials, especially those coming from the field of nuclear reactor technology, these materials are shielded from one another by forcibly beamed neutrons, depending on the job, material and condition, for example when changing and/or testing and for transport and/or storage, to prevent further nuclear reactions. To achieve the desired neutron absorption, absorber elements in the form of various types of shafts, canisters, tubes or similar configurations are usually produced that surround an object emitting neutrons and thus shield it. The use of such absorber elements permits compact storage of elements that give off neutrons, especially fuel elements from nuclear power plants, for example.

A storage rack for fuel elements is known from EP 0 385 187 A1 in which absorber sheets form a number of shafts that surround the burning element over its entire length. These absorber elements are shafts or tubes made of a material that absorbs neutrons, for example, boron steel, a high-grade steel with 1% to 2% boron. Apart from the necessary production expense, these absorber elements are extremely cost-intensive and their effectiveness is limited due to the limited proportion of boron. In an attempt to increase the proportion of boron, the deposition of a boron-nickel alloy was studied. The proportion of boron can be raised to 8%, but the costs also increase by a factor of 10, so such tubes cannot be used economically.

For other jobs, for example transport and/or storage of radioactive materials, methods are used in which nickel coatings are deposited on the metal surfaces of containers.

US-PS 4 218 622 describes a composite absorber element which has a thin carrier film or a thin carrier sheet to which a polymer matrix is applied that has boron carbide particles embedded in it. Preferably, fiberglass-reinforced polymer is used as the material for the carrier film or carrier sheet. The boron carbide particles are distributed evenly on the surface of the polymer matrix, with a boron concentration of up to 0.1 g/cm<sup>2</sup>. When the composite absorber part is used in a fuel element storage rack, this absorber element has a thickness of up to 7 mm, is designed in the form of a film or sheet and is suspended between an inner wall and an outer wall. Whether homogeneous distribution of the boron carbide particles arranged on the surface of the polymer matrix is guaranteed over a longer period of time, especially if there is friction on the surface, cannot be inferred from US-PS 4 218 622.

EP 0 016 252 A1 describes a method of producing an absorber element that absorbs neutrons. In this method, boron carbide is applied to a substrate along with a metal substance by means of plasma spraying,

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and the boron carbide is embedded in a matrix made of a metal substance. The method is also carried out so as to avoid oxidation of the boron. The absorber element produced in this way should be stable vis-à-vis a liquid medium like what exists in a fuel element storage basin, for example. The thickness of the layer of metal and boron carbide applied by means of plasma spraying is at least 500 µm. The proportion of boron carbide is roughly 50% by volume. Aluminum, copper and stainless steel can be considered for the metal substance, and the substrate contains the same metal substance as the sprayed-on layer. A relatively thick coating of boron carbide is necessary for effective neutron absorption; the thickness of the layer is 3 to 6 mm, in particular.

It is known from DE-AS 1 037 302 and DE 2 361 363 how to provide tubes, especially tin cans, with absorber material on their outer surface by electrolytic methods to protect them from radioactive radiation. There is no information on the engineering processes and equipment for technical implementation of physical-chemical changes in status and conversion of materials for applying the absorber materials that can be obtained from DE-AS 1 037 302 and DE 2 361 363.

Methods of producing shielding elements are known from EP 0 055 679 A2 in which boron carbide is either placed on the surface of the shielding element in a plasma-coating method or after electrolytic or chemical preliminary nickel plating of the shielding element, boron carbide is sprinkled in powder form on the surface and the shielding element is then nickel-plated electrolytically or chemically afterward. In this method, only small amounts of boron carbide, on the order of 20% by weight in relation to the nickel, are applied to the surface. Very heavy coatings are therefore needed, so these previously known methods are not economical. In practice, these methods are not further used, since they cannot be specifically engineered. Applying powder to a surface by sprinkling is not a measure that guarantees secure industrial production.

All the previously known methods and the shielding elements produced by them can be regarded as uneconomical in terms of high production costs and a large expenditure of materials. Moreover, the variability of the form of the shielding element and the expansion of potential uses are limited.

Production of boron steel is extremely expensive. The steel is smelted and boron is enriched by expensive methods up to a valence of 10 and mixed with the smelted steel. This yields boron steel with 1.1% to 1.4% boron by weight. This steel is very hard to work with, is extremely brittle and is difficult to solder. Shielding elements made from it are extremely heavy with average absorption properties. For example, inner storage containers, such as baskets for interim storage of fuel elements are known that are made of boron steel and weigh approximately 10 tons.

Starting from the previously known state of the art, the problem of this invention is to specify a method of producing a coating or shielding elements to absorb the neutrons created in a nuclear reaction of radioactive materials that is economical and easy to use, which increases the effectiveness of the absorption, permits greater variability in terms of the basic materials and the shape of the shielding elements and especially permits production of lighter shielding elements that have at least the same absorption qualities.

A method of producing a coating to absorb the neutrons created in a nuclear reaction of radioactive materials is proposed for the technical solution of this problem in which at least part of a shielding element composed of a basic material is provided with a boron-nickel layer on its predetermined surfaces in a dispersion bath containing boron, and during the coating process a relative movement is produced between the surface to be coated and the dispersion bath, at least for a time.

Surprisingly, it has been shown that the design of a boron-nickel coating in a dispersion bath with relative movement for a time between the surface to be coated and the dispersion bath gives very good results. In contrast to the embedding done in the past, the boron can be inserted into the nickel matrix in magnitudes >20% by volume or even  $\geq 40\%$  by volume. The boron can be contained in the dispersion as boron carbide (B<sub>4</sub>C) or, according to one especially advantageous proposal in the invention, as boron in elemental form. When elemental boron is used, even more boron can be embedded.

Thus, because of the high embedding rates, there is much greater effectiveness. The absorption layers are on the order of 350 to 500  $\mu$ m, which is extremely thin. Moreover, one special advantage is the method's independence from the basic material. It is an advantage that inorganic basic material can be used, for example steel, titanium, copper, nickel and the like. Despite its organic character and hence susceptibility to neutron radiation, carbon fiber material can be considered as a basic material. Carbon fiber material has the special advantage that the absorption element can be produced by galvanizing technology.

The invention also offers the possibility of making the shielding element in the finished state or in individual parts. Because of its independence from the basic material, materials that are very easy to work with can be used. On the other hand, very complicated forms of shielding elements, containers, baskets and the like can also be prefabricated completely and then coated according to the invention.

Because of the high embedding rate, the shielding is extremely effective, so the coatings can be extremely thin. Thus, weight savings up to 80% compared to shielding elements that can be produced by the conventional methods are possible. The former 10-ton inner storage containers (baskets) now used in the so-called Castor Program for storing fuel elements can be produced by the method in the invention on the order of 2.5 to 3 tons now.

The basic material can be prefabricated as a finished part or as individual parts, so that finished shielding elements can be made from individual parts. They are coated either chemically or electrolytically in the dispersion bath.

The relative movement between the surface to be coated and the dispersion bath can be brought about, for example, by moving the element to be coated in the dispersion bath. As is known, elements like boron can be constituted so that recirculating or pumping the dispersion is not economically possible in practice. Any recirculating or pumping unit would be worn out in a short period of time. Nevertheless, relative movement, on one hand, achieves continuously good mixing or repeated mixing of the dispersion, and on the other hand, directly taking the dispersion to the surface to be coated. Besides moving the element, the whole coating system can also be moved for the purpose of producing the relative movement. Thus, coating can conceivably take place in a type of drum, for example.

It is a special advantage proposed with the invention that the surface to be coated is arranged pointing upward in the dispersion bath. This means that the surface to be coated is arranged in the dispersion bath in such a way that because of the force of gravity, the particles found in the dispersion fall on the surface. This arrangement in the invention, especially in combination with the production of relative movement between the surface and the dispersion bath from time to time, gives outstanding coating results.

The invention offers the special advantage that the coating method is carried out in a glass tub. This guarantees special purity of the dispersion bath.

The invention provides a simple, economical and very effective method of producing shielding elements for neutron absorption, which makes it possible to produce shielding elements independent of the basic material that are much lighter than the known shielding elements with comparable absorption effects.

The invention also concerns shielding elements produced by the method described. They are characterized by the fact that they have a boron-nickel coating, with a proportion of boron in elemental form or boron carbide greater than 20% by volume or 40% by volume. The thickness of the coating is 350 to 500  $\mu$ m, and the coating is done on an inorganic basic material such as steel, titanium, copper or the like. The process is chemical or electrolytic. The shielding element can be coated in finished form or can be put together from individually coated parts.

In one experiment, conventional steel plates were coated electrolytically in a nickel/boron carbide dispersion bath. The plates were all turned every half hour in the bath and moved up and down from time to time in order to produce a relative movement between the surface and the dispersion bath, on one hand,

and to arrange the surface to be coated face up in the bath, on the other. Boron carbide in the range of 40% by volume was able to be embedded in the nickel matrix, as subsequent analyses revealed.